Interactive comment on “Trajectory-based source area analysis of atmospheric fine particles, SO$_2$, NO$_x$ and O$_3$ for the SMEAR II station in Finland in 1996–2008” by M. Hulkkonen et al.

Anonymous Referee #2

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Overview:

This article describe a method on how to identify how spatial distribution of air mass transport influence measured concentrations of different atmospheric trace constituents, including NO, O3, SO2 and particulate matter. The approach relies on statistical treatment of a combination of 13 years of hourly calculated 96h backward air mass trajectories that together with atmospheric observational data observations performed at Hyytiälä, provide information on what relative concentration can be measured at the receptor after crossing a certain grid area. The information is further used to study how the relative influence of different source areas has changed over the studied period.
Major comments:

In the abstract it is stated that the study utilize a method “...for characterizing the source areas of atmospheric SO2, NOx, O3 and aerosol particles of different size modes from the perspective of a Finnish measurement station located in Hyytiälä”

I do not agree on this. Provision of such results based on the current method would only work for a completely inert compound with no sinks combined with overall air flow conditions that would diminish the shading effect of large point sources; i.e. what this study shows is not the source areas of different compounds per se. Instead the study provide information on what measurable concentration of trace constituents an air mass contains after crossing a certain grid cell or area. Although this to some degree naturally also reflects the actual source areas, the method used cannot provide any absolute information of the source area distribution. This is due to the fact that the atmospheric lifetimes are not taken into account and as a result to the shading effect of strong point sources. However, as a method to describe flow conditions that contribute to the observed concentration at the receptor, the method seems good enough. As the manuscript is written now, a confusing terminology is used, which makes it somewhat difficult to understand what kind of information the authors are trying to provide with respect to source areas. As example, on page 1656, lines 5-7 it is stated:

“The approach applied in our study allows us to recognize the relative contribution of each grid cell to the concentrations measured at Hyytiälä”

This cannot be true since the utilized approach never can provide this information. A few lines below however, the authors conclude that:

“...the relative impact that different areas potentially have on measured values in Hyytiälä.”

which is somewhat closer to the truth although I would like to rewrite it as

“...the relative impact that air mass transport over different areas have on measured
values . . .”.

This in turn indicates a combination of source strength in that grid as well as the sink strength downwind of it. Then of course it may be discussed if this information is at all relevant depending on atmospheric lifetime and distance from the receptor. If the lifetime is short (e.g. as for nuclei mode particles and NOx) emissions far from the receptor may when the air arrives from distant sources have no significant contribution to observed concentrations. A more direct approach would include scaling to sinks/lifetime, but this of course introduces several other problems that will be hard to address.

Thus, clarifying the terminology used and making changes similar to the suggestions above where else applicable would aid in the readers interpretation of what information the method actually does provide. For example, the authors use “relative contribution” at several instances. This appears to be a misleading and overly qualitative and quantitative interpretation of the results as the method cannot provide this kind of information, especially not for short lived compounds that are emitted far from the receptor. Nevertheless, when reading the manuscript, it seem clear to me that the authors are aware of the drawbacks of the methods (i.e. last paragraph, section 2.2.1), which in principle means that many concerns regarding the manuscript at least to a certain degree are caused by misleading terminology applied by the authors. In many cases it further seems so that the authors “boil down” the result from the analysis to a division of air mass transport within different sectors, which is completely appropriate given the presented approach and method. Thus, from the perspective of air mass climatology and its relation to observed concentration at a certain receptor, the study/method seems suitable. Especially if meteorological parameters along transport also would be considered. As a way of mapping point sources/source distribution it is however less convincing. Based on this I do suggest a minor revision where the authors better and more clearly provide information regarding what the calculations of their RSCF actually resembles, how it may be used and what improvements that would be necessary to take the method one step further.
Minor comments:

In the RSCF plots per month and year, to what number is the maps normalized? Annual maximum? Monthly maximum? This is not clear.

P. 1657, line 27: “groud”→”ground”

Page 1658, lines 1-3: 1996 is included in the study, but not in the trajectory calculations!? I guess the reasons for having two datasets is that the FNL archive ends in 2007. Please clarify/revise how and for which years the datasets were used. Why partially overlapping?

Page 1664, line 15: suggest rephrasing

Page 1666, line 14-15: As with nucleation mode particles, speaking about source areas in terms of ozone does not make sense. Ozone is formed in situ in the atmosphere based on availability and proportions of both NOx and VOC (and solar radiation of course).

Section 4.2.1: How much has the annual average of SO2 observed at the receptor changed during the studied period?

Think figure 5 can be omitted as the information is already provided in figure 3-4 and 6